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A quantum-mechanical master equation treatment of the dynamical Stark effect

H J Carmichael† and D F Walls

School of Science, University of Waikato, Hamilton, New Zealand

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Abstract. A fully quantum-mechanical treatment of the dynamical Stark effect for resonant interactions is presented. The description begins with an operator master equation for the atom plus incident field. Reduced atomic matrix elements are derived for arbitrary field strengths. First- and second-order correlation functions in the scattered field are also obtained and discussed in relation to the scattered spectrum and intensity-fluctuation measurements. Our formalism has the appealing feature that all this information is readily available from the one set of four coupled equations. The deficiencies in both the one-photon approximation and the semiclassical perspective are established in a natural and transparent fashion.

1. Introduction

The subject of resonance fluorescence is a relatively old one, receiving classic treatment by Weisskopf and Wigner (1930, 1933) and later by Heitler (1954). These early calculations were based on perturbative techniques and hence restricted to weak scattering situations where the atom remains close to its ground state and behaves to a good approximation as the electron oscillator of the Lorentzian theory. An increased incident intensity alters the behaviour quite considerably, however, as the atom is then driven significantly from its ground state. The phenomenon of resonance fluorescence then exhibits new features associated with a behaviour which has come to be known as the dynamical or AC Stark effect. Here the quantum nature of the scattering source makes its mark, and in particular we find a quantum statistical component in the spectrum.

Renewed interest has recently arisen in resonance fluorescence in relation to this nonlinear region. This has stemmed from proposals (Stroud and Jaynes 1970) that here there may be a test for QED as against the neoclassical theory for an atom reacting with the radiation field (Jaynes and Cummings 1963, Crisp and Jaynes 1969, Stroud and Jaynes 1970). Of particular interest for example is the long-time behaviour of the scattered spectrum. This is predicted in the neoclassical theory to reduce to a sharp line at the incident frequency, contrary to the quantum-mechanical expectation (Newstein 1968, Mollow 1969, Stroud 1971, 1973). In response to these proposals and the subsequent experimental verification of a three-peaked spectrum well into atomic

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saturation (Schuda *et al* 1974, Walther 1975, Wu *et al* 1975, Hartig *et al* 1976) a number of theoretical discussions have appeared (Chang and Stehle 1971, Gush and Gush 1972, Smithers and Freedhoff 1974, 1975, Hassan and Bullough 1975, Mollow 1975a, b, Carmichael and Walls 1975a, Swain 1975, Cohen-Tannoudji 1975, 1976). Although in the course of this 'dialogue' differences have arisen in the various predictions, there appears now to be emerging a general agreement on the form of the scattered spectrum for intense fields and its long-time behaviour.

We present here a fully quantum-mechanical analysis of the resonance fluorescence problem for a two-level atom in a resonant field. Our fundamental starting point is an operator master equation in the Markoffian approximation. While this approximation has been criticized by some authors we may now rely on its recent verification for this specific application by Mollow (1975) and Epstein (1975). From our point of view we find that it has considerable tutorial advantages over those treatments which begin directly with Schrödinger's equation. The various features of atomic dynamics and scattered spectrum are readily available from an essentially simple analysis and in a language indicating a stochastic process whose statistics have a purely quantum-mechanical origin. This serves as an uncomplicated and worthwhile perspective on the problem. Moreover we find that the second-order correlation function may be extracted with little extra work from the same equations that give atomic matrix elements and the first-order correlation function. This is a feature which has not previously been discussed in the literature and has considerable significance in that it gives rise to an interesting proposal for the measurement of spectral detail by optical homodyne or heterodyne spectroscopy.

In §2 we outline the formal apparatus underlying our approach. Atomic dynamics are then investigated in §3 in terms of solutions to optical Bloch equations and the semiclassical form of the field is indicated. The first-order correlation function and scattered spectrum are obtained in §4 and the one-photon approximation demonstrated in §5. The second-order correlation function is derived in §6 and proposed as a basis for a new measurement of linewidths and frequency splitting.

2. Formal apparatus

The phenomenon of resonance fluorescence arises with the illumination of an atomic dipole transition by resonant radiation and appears as scattering from the incident beam into other modes of the radiation field. In subsequent sections we present results on both atomic dynamics and correlations in the scattered field. Here we formulate our approach and outline the formal apparatus employed to meet these ends.

We consider a two-level atom coupled at resonance to a highly populated field mode and simultaneously interacting with the vacuum. Our treatment begins with the perspective which sees here an open system S interacting with a thermal reservoir R at zero temperature. Dividing the radiation field into incident and scattered components, $E_I(r, t)$ and $E_S(r, t)$, with

$$\begin{aligned} E_I(r, t) &= i \left(\frac{\hbar \omega_0}{2\mathcal{E}_0 V} \right)^{1/2} \hat{e}_0 (a(t) e^{ik_0 \cdot r} - a^\dagger(t) e^{-ik_0 \cdot r}) \\ E_S(r, t) &= i \sum_{\mathbf{k}, \lambda} \left(\frac{\hbar \omega_k}{2\mathcal{E}_0 V} \right)^{1/2} \hat{e}_{\mathbf{k}, \lambda} (b_{\mathbf{k}, \lambda}(t) e^{i\mathbf{k} \cdot \mathbf{r}} - b_{\mathbf{k}, \lambda}^\dagger(t) e^{-i\mathbf{k} \cdot \mathbf{r}}) \end{aligned} \quad (2.1)$$

we then write the Hamiltonian for the composite system $S \oplus R$ in the form

$$H = H_S + H_R + H_{SR} \quad (2.2)$$

with

$$\begin{aligned} H_S &= \hbar\omega_0 a^\dagger a + \frac{1}{2}\hbar\omega_0 \sigma_z + \hbar(\kappa_0 a^\dagger \sigma_- + \kappa_0^* a \sigma_+) \\ H_R &= \sum_{\mathbf{k}, \lambda} \hbar\omega_{\mathbf{k}} b_{\mathbf{k}, \lambda}^\dagger b_{\mathbf{k}, \lambda} \quad H_{SR} = \sum_{\mathbf{k}, \lambda} \hbar(\kappa_{\mathbf{k}, \lambda} b_{\mathbf{k}, \lambda}^\dagger \sigma_- + \kappa_{\mathbf{k}, \lambda}^* b_{\mathbf{k}, \lambda} \sigma_+). \end{aligned} \quad (2.3)$$

$a^\dagger$ and a create and annihilate photons in the incident beam with frequency ω_0 , wavevector $\mathbf{k}_0$ and polarization $\hat{\mathbf{e}}_0$; $b_{\mathbf{k}, \lambda}^\dagger$ and $b_{\mathbf{k}, \lambda}$ fulfil a similar purpose for the vacuum mode of frequency $\omega_{\mathbf{k}}$, wavevector $\mathbf{k}$ and polarization $\hat{\mathbf{e}}_{\mathbf{k}, \lambda}$; σ_z , σ_+ and σ_- are the usual atomic pseudo spin operators and κ_0 and $\kappa_{\mathbf{k}, \lambda}$ are coupling constants for atomic dipole interaction of frequency ω_0 , $\omega_{\mathbf{k}}$ and polarization $\hat{\mathbf{e}}_0$, $\hat{\mathbf{e}}_{\mathbf{k}, \lambda}$. The interaction between the radiation field and the atom has been written in the electric dipole and rotating-wave approximations.

The derivation of a master equation for the reduced density operator ρ of S is well known and under the Markoffian assumption, the familiar procedures (see, for example, Agarwal 1973) lead to the form

$$\frac{d\rho}{dt} = \frac{1}{i\hbar} [H_S, \rho] + \frac{\gamma}{2} (2\sigma_- \rho \sigma_+ - \rho \sigma_+ \sigma_- - \sigma_+ \sigma_- \rho) \quad (2.4)$$

where γ corresponds to the Einstein A coefficient. This we may express formally as

$$\frac{d\rho}{dt} = \mathcal{L}\rho \quad (2.5)$$

with the generalized Liouvillian $\mathcal{L}$ defined by

$$\mathcal{L}O = \frac{1}{i\hbar} [H_S, O] + \frac{\gamma}{2} (2\sigma_- \rho \sigma_+ - \rho \sigma_+ \sigma_- - \sigma_+ \sigma_- \rho) \quad (2.6)$$

O being an arbitrary operator.

It is convenient that we work in the energy representation corresponding to the Hamiltonian of S . For the familiar eigenstates of H_S we have

$$\begin{aligned} H_S |E_n^2\rangle &= \hbar[\omega_0(n + \frac{1}{2}) + \kappa(n + 1)^{1/2}] |E_n^2\rangle \\ H_S |E_n^1\rangle &= \hbar[\omega_0(n + \frac{1}{2}) - \kappa(n + 1)^{1/2}] |E_n^1\rangle \end{aligned} \quad (2.7)$$

where

$$\begin{aligned} |E_n^2\rangle &= \frac{1}{\sqrt{2}}(|n, +\rangle + |n + 1, -\rangle) \\ |E_n^1\rangle &= \frac{1}{\sqrt{2}}(|n, +\rangle - |n + 1, -\rangle) \end{aligned} \quad (2.8)$$

$|n\rangle$ are the Fock states and $|+\rangle$ and $|-\rangle$ the upper and lower atomic states. Within this representation, equation (2.4) takes the form

$$\frac{d\rho_{n,\eta;m,\xi}}{dt} = -\frac{i}{\hbar} (E_n^\eta - E_m^\xi) \rho_{n,\eta;m,\xi} + (-1)^{n+\xi} \frac{\gamma}{4} \sum_{\mu\nu} \rho_{n+1,\mu;m+1,\nu} - \frac{\gamma}{4} \sum_{\nu} (\rho_{n,\eta;m,\nu} + \rho_{n,\nu;m,\xi}) \quad (2.9)$$

where

$$\rho_{n,\eta;\xi} = \langle E_n^\eta | \rho | E_n^\xi \rangle \quad \eta = 1, 2 \quad \xi = 1, 2. \quad (2.10)$$

We have here an infinite set of coupled equations indicating the successive scattering of photons from the incident beam and the consequent eventual decay of all matrix elements to those of lower photon number. It will nevertheless be unnecessary, so far as the fluorescent field itself is concerned, to solve such a complex system. We may put the detailed dynamical structure aside and deal only with atomic matrix elements summed over the incident field. We define $\rho_{\xi,\eta}$ by

$$\rho_{\xi,\eta} = \sum_n \rho_{n,\xi;n,\eta}. \quad (2.11)$$

The scattered field may be expressed in terms of the atomic source operators for times $\omega_0 t \gg 1$ (Agarwal 1974, Ackerhalt and Eberly 1974, Kimble and Mandel 1975a, b, Saunders *et al* 1975) by

$$E_S(r, t) = E_S^{(+)}(r, t) + E_S^{(-)}(r, t) \quad (2.12)$$

with

$$E_S^{(+)}(r, t) = \mathcal{E}_S^{(+)}(r, t) - \frac{\omega_0^2}{4\pi\epsilon_0 c^2 r} \left(\boldsymbol{\mu} \times \frac{\mathbf{r}}{r} \right) \times \frac{\mathbf{r}}{r} \sigma_- \left(t - \frac{r}{c} \right). \quad (2.13)$$

$\mathcal{E}_S^{(+)}(r, t)$ describes a freely propagating field and the retarded source term is simply the retarded field generated by a point dipole (Landau and Lifshitz 1962); $\boldsymbol{\mu}$ is the atomic dipole moment. With this result we may then clearly obtain field correlations from the dynamical information within equation (2.9). In particular, for the first- and second-order correlation functions given by

$$G^{(1)}(r, t; r, t + \tau) = \langle E_S^{(-)}(r, t) E_S^{(+)}(r, t + \tau) \rangle \quad (2.14)$$

$$G^{(2)}(r, t; r, t + \tau) = \langle E_S^{(-)}(r, t) E_S^{(-)}(r, t + \tau) E_S^{(+)}(r, t + \tau) E_S^{(+)}(r, t) \rangle \quad (2.15)$$

we may write respectively

$$G^{(1)}(r, t; r, t + \tau) = I_0(r) \langle \sigma_+(\hat{t}) \sigma_-(\hat{t} + \tau) \rangle \quad (2.16)$$

$$G^{(2)}(r, t; r, t + \tau) = I_0(r)^2 \langle \sigma_+(\hat{t}) \sigma_+(\hat{t} + \tau) \sigma_-(\hat{t} + \tau) \sigma_-(\hat{t}) \rangle \quad (2.17)$$

where $\hat{t} = t - r/c$ and

$$I_0(r) = \left[\frac{\omega_0^2}{4\pi\epsilon_0 c^2 r} \left(\boldsymbol{\mu} \times \frac{\mathbf{r}}{r} \right) \times \frac{\mathbf{r}}{r} \right]^2 \quad (2.18)$$

is the intensity detected at position r at the retarded time $\hat{t} = 0$. Our concern is then with the evaluation of the two-time averages $\langle \sigma_+(\hat{t}) \sigma_-(\hat{t} + \tau) \rangle$ and $\langle \sigma_+(\hat{t}) \sigma_+(\hat{t} + \tau) \sigma_-(\hat{t} + \tau) \sigma_-(\hat{t}) \rangle$. For this purpose the quantum regression theorem of Lax is available (Lax 1967).

The validity of the quantum regression theorem in this problem has been established within the rotating-wave approximation (see Mollow 1975a, b and references therein). Thus we find for $\tau \geq 0$ the expressions

$$\langle \sigma_+(\hat{t}) \sigma_-(\hat{t} + \tau) \rangle = \text{Tr}_S [\sigma_- e^{\mathcal{L}\tau} \rho(\hat{t}) \sigma_+] \quad (2.19)$$

and

$$\langle \sigma_+(\hat{t}) \sigma_+(\hat{t} + \tau) \sigma_-(\hat{t} + \tau) \sigma_-(\hat{t}) \rangle = \text{Tr}_S [\sigma_+ \sigma_- e^{\mathcal{L}\tau} (\sigma_- \rho(\hat{t}) \sigma_+)]. \quad (2.20)$$

The determination of the matrix elements (2.11) and averages (2.19) and (2.20) is readily made from the solution of a simple set of four coupled equations. We merely assume an incident field of high intensity, possessing a sharply peaked photon-number distribution. We may for example take a coherent state corresponding to laser radiation

$$|\alpha\rangle = \exp(-\frac{1}{2}|\alpha|^2) \sum_n \frac{\alpha^n}{(n!)^{1/2}} |n\rangle. \quad (2.21)$$

Here $\bar{n} = |\alpha|^2$, which for the Poisson distribution also gives the variance. Then for $\bar{n} \sim 10^8$ a fractional change in photon number of 10^{-4} only arises over one standard deviation. Combined with a parametric approximation the result of this circumstance is that in summing matrix elements over the field, the factor $-i(E_n^\eta - E_m^\xi)/\hbar$ in equation (2.9) may be taken through the summation as $-i(E_n^\eta - E_n^\xi)/\hbar$. We find for $\rho_{\eta,\xi}$ the coupled equations

$$\frac{d}{d\tau} \begin{pmatrix} \rho_{22} \\ \rho_{11} \\ \rho_{21} \\ \rho_{12} \end{pmatrix} = \begin{pmatrix} -\frac{1}{4}\gamma & \frac{1}{4}\gamma & 0 & 0 \\ \frac{1}{4}\gamma & -\frac{1}{4}\gamma & 0 & 0 \\ -\frac{1}{2}\gamma & -\frac{1}{2}\gamma & -(\frac{3}{4}\gamma + 2i\bar{n}^{1/2}\kappa) & -\frac{1}{4}\gamma \\ -\frac{1}{2}\gamma & -\frac{1}{2}\gamma & -\frac{1}{4}\gamma & -(\frac{3}{4}\gamma - 2i\bar{n}^{1/2}\kappa) \end{pmatrix} \begin{pmatrix} \rho_{22} \\ \rho_{11} \\ \rho_{21} \\ \rho_{12} \end{pmatrix}. \quad (2.22)$$

Essentially the same set of equations may be used in solving for the averages (2.19) and (2.20). We may define respectively the two operators $\Sigma(\tau)$ and $\Pi(\tau)$ by

$$\begin{aligned} \Sigma(\tau) &= e^{\mathcal{L}\tau} \Sigma(0) \\ \Sigma(0) &= \rho(\hat{t})\sigma_+ \end{aligned} \quad (2.23)$$

and

$$\begin{aligned} \Pi(\tau) &= e^{\mathcal{L}\tau} \Pi(0) \\ \Pi(0) &= \sigma_- \rho(\hat{t})\sigma_+. \end{aligned} \quad (2.24)$$

It then clearly follows that we may write

$$\begin{aligned} \frac{d\Sigma(\tau)}{d\tau} &= \mathcal{L}\Sigma(\tau) \\ \frac{d\Pi(\tau)}{d\tau} &= \mathcal{L}\Pi(\tau) \end{aligned} \quad (2.25)$$

and that matrix elements of these operators are consequently determined by equations formally equivalent to equation (2.9). After a transformation $\Sigma_{\eta,\xi}(\tau) = \hat{\Sigma}_{\eta,\xi}(\tau)e^{-i\omega_0\tau}$, the matrix elements of these operator equations (2.25) reduce to equations which are equivalent to equations (2.22) under the same conditions which lead to equations (2.22). Moreover, evaluating the respective traces, (equations (2.19) and (2.20)) yields

$$\langle \sigma_+(\hat{t})\sigma_-(\hat{t} + \tau) \rangle = \frac{1}{2}(\Sigma_{22}(\tau) - \Sigma_{11}(\tau)) - \frac{1}{2}(\Sigma_{21}(\tau) - \Sigma_{12}(\tau)) \quad (2.26)$$

and

$$\langle \sigma_+(\hat{t})\sigma_+(\hat{t} + \tau)\sigma_-(\hat{t} + \tau)\sigma_-(\hat{t}) \rangle = \frac{1}{2}(\Pi_{22}(\tau) + \Pi_{11}(\tau)) + \frac{1}{2}(\Pi_{21} + \Pi_{12}(\tau)) \quad (2.27)$$

where the matrix elements $\Sigma_{\eta,\xi}(\tau)$ and $\Pi_{\eta,\xi}(\tau)$ are respectively defined by

$$\Sigma_{\eta,\xi}(\tau) = \Sigma_n \langle E_{n+1}^\eta | \Sigma(\tau) | E_n^\xi \rangle \quad (2.28)$$

and

$$\Pi_{n,\xi}(\tau) = \Sigma_n \langle E_n^\eta | \Pi(\tau) | E_n^\xi \rangle. \quad (2.29)$$

We therefore simply require the solutions to a single set of four equations (equations (2.22)) in order to evaluate both atomic matrix elements and first- and second-order field correlations.

3. Atomic dynamics and the semiclassical field

The coupled equations (2.22) are easily solved for atomic matrix elements. Defining the vector $\tilde{\rho}$

$$\tilde{\rho} = \frac{1}{2} \begin{pmatrix} \rho_{22} + \rho_{11} \\ \rho_{22} - \rho_{11} \\ \rho_{21} + \rho_{12} \\ \rho_{21} - \rho_{12} \end{pmatrix} \quad (3.1)$$

we find the formal solution

$$\tilde{\rho}(t) = \mathbf{S} \exp(\Lambda t) \mathbf{S}^{-1} \tilde{\rho}(0) \quad (3.2)$$

where

$$\mathbf{S} = \begin{pmatrix} S_1 & 0 & 0 & 0 \\ 0 & S_2 & 0 & 0 \\ -\frac{\gamma^2}{\gamma^2 + 8\bar{n}\kappa^2} S_1 & 0 & S_3 & \frac{-2i\bar{n}^{1/2}\kappa}{\frac{1}{4}\gamma - \Omega} S_4 \\ \frac{2i\bar{n}^{1/2}\kappa}{\frac{1}{2}\gamma} \frac{\gamma^2}{\gamma^2 + 8\bar{n}\kappa^2} S_1 & 0 & \frac{2i\bar{n}^{1/2}\kappa}{\frac{1}{4}\gamma - \Omega} S_3 & S_4 \end{pmatrix} \quad (3.3)$$

and S_1, S_2, S_3 and S_4 are arbitrary constants.

Λ is the 4×4 diagonal matrix

$$\Lambda = D(0, -\frac{1}{2}\gamma, -\frac{3}{4}\gamma + \Omega, -\frac{3}{4}\gamma - \Omega) \quad (3.4)$$

with

$$\Omega = [(\frac{1}{4}\gamma)^2 - 4\bar{n}\kappa^2]^{1/2}. \quad (3.5)$$

If we begin with an atom in its lower state the solutions, written in terms of the energy representation of the free atom (using equations (2.8) and (2.10)) are

$$\rho_{+,+}(t) = \frac{4\bar{n}\kappa^2}{\gamma^2 + 8\bar{n}\kappa^2} \left[1 - e^{-3\gamma t/4} \left(\cosh \Omega t + \frac{\frac{3}{4}\gamma}{\Omega} \sinh \Omega t \right) \right] \quad (3.6)$$

$$\begin{aligned} \rho_{+,-}(t) = & -2i\bar{n}^{1/2}\kappa \left\{ \frac{\gamma}{\gamma^2 + 8\bar{n}\kappa^2} \left[1 - e^{-3\gamma t/4} \left(\cosh \Omega t + \frac{\frac{3}{4}\gamma}{\Omega} \sinh \Omega t \right) \right] \right. \\ & \left. + \frac{1}{2} e^{-3\gamma t/4} \frac{\sinh \Omega t}{\Omega} \right\} \end{aligned} \quad (3.7)$$

where we have defined $\rho_{+,+}(t)$ and $\rho_{+,-}(t)$ by

$$\begin{aligned}\rho_{+,+}(t) &= \sum_n \langle n, + | \rho(t) | n, + \rangle \\ \rho_{+,-}(t) &= \sum_n \langle n, + | \rho(t) | n+1, - \rangle.\end{aligned}\quad (3.8)$$

Naturally

$$\begin{aligned}\rho_{-,-}(t) &= 1 - \rho_{+,+}(t) \\ \rho_{-,+}(t) &= \rho_{+,-}(t)^*.\end{aligned}\quad (3.9)$$

Similar solutions follow for an initially excited atom and include an additional contribution accounting, in the weak-coupling limit, for normal spontaneous emission. These solutions are

$$\begin{aligned}\rho_{+,+}(t) &= \frac{4\bar{n}\kappa^2}{\gamma^2 + 8\bar{n}\kappa^2} \left[1 - e^{-3\gamma t/4} \left(\cosh \Omega t + \frac{3}{4} \frac{\gamma}{\Omega} \sinh \Omega t \right) \right] \\ &\quad + e^{-3\gamma t/4} \left(\cosh \Omega t - \frac{1}{4} \frac{\gamma}{\Omega} \sinh \Omega t \right)\end{aligned}\quad (3.10)$$

$$\begin{aligned}\rho_{+,-}(t) &= -2i\bar{n}^{1/2}\kappa \left\{ \frac{\gamma}{\gamma^2 + 8\bar{n}\kappa^2} \left[1 - e^{-3\gamma t/4} \left(\cosh \Omega t + \frac{3}{4} \frac{\gamma}{\Omega} \sinh \Omega t \right) \right] \right. \\ &\quad \left. - \frac{1}{2} e^{-3\gamma t/4} \frac{\sinh \Omega t}{\Omega} \right\}.\end{aligned}\quad (3.11)$$

Similar solutions to the semiclassical Bloch equations for a spin $\frac{1}{2}$ system in a combination of static and RF magnetic fields were first given by Torrey (1949).

In these solutions we see the dynamics separating into an initial transient regime, followed by a saturation steady state. For weak coupling, the saturated atom settles close to its lower level and we expect the behaviour of a classical electron oscillator. With increased incident intensity, however, we find the saturation steady state moves into the nonlinear region

$$\rho_{+,+}^{\text{ss}} = \frac{4\bar{n}\kappa^2}{\gamma^2 + 8\bar{n}\kappa^2}.\quad (3.12)$$

For very intense illumination, a limit is reached midway between the upper and lower levels

$$\lim_{\bar{n} \rightarrow \infty} \rho_{+,+}^{\text{ss}} = \frac{1}{2}.\quad (3.13)$$

Quantum fluctuations may therefore be expected to become important with intense illumination, while remaining of no consequence for weak scattering.

To appreciate the predictions of the semiclassical perspective on the scattered field, we calculate the mean field which from equation (2.13) reads

$$\langle E_s^{(+)}(\mathbf{r}, t) \rangle = - \frac{\omega_0^2}{4\pi\epsilon_0 c^2 r} \left(\boldsymbol{\mu} \times \frac{\mathbf{r}}{r} \right) \times \frac{\mathbf{r}}{r} \langle \sigma_-(\hat{t}) \rangle\quad (3.14)$$

where the average $\langle \sigma_-(\hat{t}) \rangle$ is given by

$$\langle \sigma_-(\hat{t}) \rangle = \sum_n \langle n, + | \rho(\hat{t}) | n, - \rangle.\quad (3.15)$$

Thus for the coherent-state field defined by equation (2.21) where $\alpha = |\alpha| e^{i\phi}$ we find for the mean field

$$\langle E_S^{(+)}(\mathbf{r}, t) \rangle = - \left[\frac{\omega_0^2}{4\pi\epsilon_0 c^2 r} \left(\boldsymbol{\mu} \times \frac{\mathbf{r}}{r} \right) \times \frac{\mathbf{r}}{r} \right] \rho_{+,-}(\hat{t}) e^{-i(\omega_0 \hat{t} + \phi)} \quad (3.16)$$

where $\rho_{+,-}(\hat{t})$ is given by equations (3.10) and (3.11) for an atom prepared in lower and upper states respectively.

Here again is the initial transient region followed by saturation. For very intense incident radiation

$$4\bar{n}\kappa^2 \gg (\frac{1}{4}\gamma)^2 \quad (3.17)$$

we may write

$$\begin{aligned} \langle E_S^{(+)}(\mathbf{r}, t) \rangle = & \left[\frac{\omega_0^2}{4\pi\epsilon_0 c^2 r} \left(\boldsymbol{\mu} \times \frac{\mathbf{r}}{r} \right) \times \frac{\mathbf{r}}{r} \right] 2i\bar{n}^{1/2}\kappa \frac{\gamma}{\gamma^2 + 8\bar{n}\kappa^2} [e^{-i(\omega_0 \hat{t} + \phi)} - \frac{1}{2} e^{-3\gamma\hat{t}/4} \\ & (\exp\{-[i(\omega_0 + 2\bar{n}^{1/2}\kappa)\hat{t} + \phi]\} + \exp\{-[i(\omega_0 - 2\bar{n}^{1/2}\kappa)\hat{t} + \phi]\})] \end{aligned} \quad (3.18)$$

which would predict semiclassically a three-component spectrum through the transient region reducing to a sharp line for long times. This is an erroneous prediction however as has been confirmed by recent experiments (Schuda *et al* 1974, Wu *et al* 1975, Walther 1975, Hartig *et al* 1976). We will see in the following section how this inadequacy arises in neglecting quantum fluctuations. For weak illumination the sharp spectrum is as expected from classical electron-oscillator theory.

4. The first-order correlation function: scattered spectrum

In this section we present solutions for the first-order correlation function (2.14) from which we derive the scattered spectrum. This spectrum is defined in terms of the probability $P(\omega, \mathbf{r}, T)$ for photon detection by a monochromatic detector during interval T . We have the result

$$P(\omega, \mathbf{r}, T) \propto \int_{r/c}^T dt_1 \int_{r/c}^T dt_2 e^{i\omega(t_2 - t_1)} G^{(1)}(\mathbf{r}, t_1; \mathbf{r}, t_2) \quad (4.1)$$

and with normalization so the integrated spectrum gives the intensity $I(\mathbf{r}, T) = G'(\mathbf{r}, t; \mathbf{r}, t)$ we then define the spectrum $I(\omega, \mathbf{r}, T)$ by

$$I(\omega, \mathbf{r}, T) = \frac{1}{2\pi} \frac{I(\mathbf{r}, T)}{\int_{r/c}^T dt I(\mathbf{r}, t)} P(\omega, \mathbf{r}, T). \quad (4.2)$$

Since

$$G^{(1)}(\mathbf{r}, t_1; \mathbf{r}, t_2) = G^{(1)}(\mathbf{r}, t_2; \mathbf{r}, t_1)^* \quad (4.3)$$

it readily follows that

$$I(\omega, \mathbf{r}, T) = \frac{1}{2\pi} \frac{I(\mathbf{r}, T)}{\int_{r/c}^T dt I(\mathbf{r}, t)} 2 \operatorname{Re} \int_{r/c}^T dt \int_0^{t-r/c} d\tau e^{i\omega\tau} G^{(1)}(\mathbf{r}, t; \mathbf{r}, t + \tau) \quad (4.4)$$

where we have introduced t and τ with $t_1 \rightarrow t$ and $\tau = t_2 - t$.

Now the first-order correlation function, as given by equations (2.16), (2.18), (2.19) and (2.26), follows from the solution of equations (2.30). This is provided by equation

(3.2) with the introduction of the factor $e^{-i\omega_0\tau}$. Thus, defining

$$\tilde{\Sigma} = \frac{1}{2} \begin{pmatrix} \Sigma_{22} + \Sigma_{11} \\ \Sigma_{22} - \Sigma_{11} \\ \Sigma_{21} + \Sigma_{12} \\ \Sigma_{21} - \Sigma_{12} \end{pmatrix} \quad (4.5)$$

we write

$$\tilde{\Sigma}(\tau) = e^{-i\omega_0\tau} \mathbf{S} \exp(\mathbf{A}\tau) \mathbf{S}^{-1} \tilde{\Sigma}(0) \quad (4.6)$$

and from equations (2.23) and (2.28) the initial vector $\tilde{\Sigma}(0)$ is given by

$$\tilde{\Sigma}(0) = \frac{1}{2} \begin{pmatrix} \rho_{-,+}(\hat{t}) \\ \rho_{+,+}(\hat{t}) \\ -\rho_{-,+}(\hat{t}) \\ -\rho_{+,+}(\hat{t}) \end{pmatrix}. \quad (4.7)$$

Due to the complexity of the solutions for atomic matrix elements and hence $\tilde{\Sigma}(0)$ a general solution would serve no purpose here. If we concern ourselves now solely with the steady state however, from equations (3.6), (3.7) and (3.9) we may define $\tilde{\Sigma}(0)$ in equation (4.7) by

$$\begin{aligned} \rho_{+,+}^{ss} &= \frac{4\bar{n}\kappa^2}{\gamma^2 + 8\bar{n}\kappa^2} \\ \rho_{-,-}^{ss} &= 1 - \frac{4\bar{n}\kappa^2}{\gamma^2 + 8\bar{n}\kappa^2} \\ \rho_{+,-}^{ss} &= -2i\bar{n}^{1/2}\kappa \frac{\gamma}{\gamma^2 + 8\bar{n}\kappa^2} \\ \rho_{-+}^{ss} &= 2i\bar{n}^{1/2}\kappa \frac{\gamma}{\gamma^2 + 8\bar{n}\kappa^2} \end{aligned} \quad (4.8)$$

and hence find

$$\begin{aligned} G_{ss}^{(1)}(\tau) &= I_0(r) \frac{4\bar{n}\kappa^2}{\gamma^2 + 8\bar{n}\kappa^2} \left[\frac{\gamma^2}{\gamma^2 + 8\bar{n}\kappa^2} e^{-i\omega_0\tau} + \frac{1}{2} \exp[-(\frac{1}{2}\gamma + i\omega_0)\tau] \right. \\ &\quad - \frac{1}{2} \left(\frac{\gamma^2}{\gamma^2 + 8\bar{n}\kappa^2} \frac{\frac{3}{4}\gamma + \Omega}{\Omega} - \frac{\frac{1}{2}\gamma}{\Omega} - \frac{1}{2} \frac{\frac{1}{4}\gamma + \Omega}{\Omega} \right) \exp\{-[(\frac{3}{4}\gamma - \Omega) + i\omega_0]\tau\} \\ &\quad \left. + \frac{1}{2} \left(\frac{\gamma^2}{\gamma^2 + 8\bar{n}\kappa^2} \frac{\frac{3}{4}\gamma - \Omega}{\Omega} - \frac{\frac{1}{2}\gamma}{\Omega} - \frac{1}{2} \frac{\frac{1}{4}\gamma - \Omega}{\Omega} \right) \exp\{-[(\frac{3}{4}\gamma + \Omega) + i\omega_0]\tau\} \right]. \end{aligned} \quad (4.9)$$

Taking the limit for $T \rightarrow \infty$, the spectrum (4.4) is given by the Fourier transform

$$I(\omega, r, \infty) = \frac{1}{2\pi} 2 \operatorname{Re} \int_0^\infty e^{i\omega\tau} G_{ss}^{(1)}(\tau). \quad (4.10)$$

Thus, in the definition (3.5) for Ω we find a threshold at

$$2\bar{n}^{1/2}\kappa = \frac{1}{4}\gamma. \quad (4.11)$$

Here the spectrum splits, changing from a single peak to a central peak plus equally spaced sidebands. For weak illumination,

$$4\bar{n}\kappa^2 \ll (\frac{1}{4}\gamma)^2 \quad (4.12)$$

equation (4.9) reduces to

$$G_{ss}^{(1)}(\tau) = I_0(r) \frac{4\bar{n}\kappa^2}{\gamma^2 + 8\bar{n}\kappa^2} e^{-i\omega_0\tau} \quad (4.13)$$

and in the strong-coupling limit (equation (3.17)) we have

$$G_{ss}^{(1)}(\tau) = \frac{I_0(r)}{2} \left(\frac{\gamma^2}{\gamma^2 + 8\bar{n}\kappa^2} e^{-i\omega_0\tau} + \frac{1}{2} \exp[-(\frac{1}{2}\gamma + i\omega_0)\tau] + \frac{1}{4} \exp\{-[\frac{1}{4}\gamma + i(\omega_0 + 2\bar{n}^{1/2}\kappa)]\tau\} + \frac{1}{4} \exp\{-[\frac{3}{4}\gamma + i(\omega_0 - 2\bar{n}^{1/2}\kappa)]\tau\} \right). \quad (4.14)$$

Thus, from equation (4.10), for weak scattering we find

$$I(\omega, r, \infty) = I_0(r) \frac{4\bar{n}\kappa^2}{\gamma^2 + 8\bar{n}\kappa^2} \delta(\omega - \omega_0) \quad (4.15)$$

and we regain the sharp spectrum predicted by the semiclassical theory. For very intense fields however, equations (4.10) and (4.15) give the results

$$I(\omega, r, \infty) = \frac{I_0(r)}{2\pi} \left(2\pi \frac{\gamma}{\gamma^2 + 8\bar{n}\kappa^2} \delta(\omega - \omega_0) + \frac{1}{2} \frac{\frac{1}{2}\gamma}{(\frac{1}{2}\gamma)^2 + (\omega - \omega_0)^2} + \frac{1}{4} \frac{\frac{3}{4}\gamma}{(\frac{3}{4}\gamma)^2 + [\omega - (\omega_0 + 2\bar{n}^{1/2}\kappa)]^2} + \frac{1}{4} \frac{\frac{3}{4}\gamma}{(\frac{3}{4}\gamma)^2 + [\omega - (\omega_0 - 2\bar{n}^{1/2}\kappa)]^2} \right). \quad (4.16)$$

Thus, added to the coherent scattering, we see three peaks arising from the quantum fluctuations, in contrast to the picture presented for the steady state by equation (3.18).

As an example of the behaviour of this first-order correlation function and spectrum in the transient region, let us now consider the time-dependent matrix elements in equation (4.7) but make a restriction to the intense field limit (3.17). An expression for the transient spectrum has not previously been obtained. For an initially excited atom equations (3.10), (3.11) and (3.9) give

$$\begin{aligned} \rho_{+,+}(\hat{t}) &= \frac{1}{2}(1 + e^{-3\gamma\hat{t}/4} \cos 2\bar{n}^{1/2}\kappa\hat{t}) \\ \rho_{-,-}(\hat{t}) &= \frac{1}{2}(1 - e^{-3\gamma\hat{t}/4} \cos 2\bar{n}^{1/2}\kappa\hat{t}) \\ \rho_{+,-}(\hat{t}) &= -\frac{1}{2}i e^{-3\gamma\hat{t}/4} \sin 2\bar{n}^{1/2}\kappa\hat{t} \\ \rho_{-,+}(\hat{t}) &= \frac{1}{2}i e^{-3\gamma\hat{t}/4} \sin 2\bar{n}^{1/2}\kappa\hat{t}. \end{aligned} \quad (4.17)$$

Introducing these into equation (4.7) we then find

$$\begin{aligned} G^{(1)}(r, t; r, t + \tau) &= \frac{1}{2}I_0 \left(\frac{1}{2} [1 + \exp(-\frac{3}{4}\gamma\hat{t}) \cos 2\bar{n}^{1/2}\kappa\hat{t}] \exp[-(\frac{1}{2}\gamma + i\omega_0)\tau] \right. \\ &\quad + \frac{1}{4} \{1 + \exp[-(\frac{3}{4}\gamma - 2i\bar{n}^{1/2}\kappa)\tau]\} \exp[-(\frac{3}{4}\gamma + i\omega_{21})\tau] \\ &\quad \left. + \frac{1}{4} \{1 + \exp[-(\frac{3}{4}\gamma + 2i\bar{n}^{1/2}\kappa)\tau]\} \exp[-(\frac{3}{4}\gamma + i\omega_{12})\tau] \right) \end{aligned} \quad (4.18)$$

where we define

$$\begin{aligned}\omega_{21} &= \omega_0 + 2\bar{n}^{1/2}\kappa \\ \omega_{12} &= \omega_0 - 2\bar{n}^{1/2}\kappa.\end{aligned}\quad (4.19)$$

We have managed the various integrals involved in equation (4.4) to obtain the time-dependent spectrum. For normalization we find

$$I(r, T) = \frac{1}{2}I_0(r)(1 + e^{-3\gamma\hat{T}/4}\cos 2\bar{n}^{1/2}\kappa\hat{T}) \quad (4.20)$$

and

$$\begin{aligned}\int_{r/c}^T dt I(r, t) &= \frac{1}{2}I_0(r)\left(\hat{T} + \frac{\frac{3}{4}\gamma}{(\frac{3}{4}\gamma)^2 + 4\bar{n}\kappa^2}(1 - e^{-3\gamma\hat{T}/4}\cos 2\bar{n}^{1/2}\kappa\hat{T})\right. \\ &\quad \left.+ \frac{2\bar{n}^{1/2}\kappa}{(\frac{3}{4}\gamma)^2 + 4\bar{n}\kappa^2}e^{-3\gamma\hat{T}/4}\sin 2\bar{n}^{1/2}\kappa\hat{T}\right) \quad (4.21)\end{aligned}$$

with $\hat{T} = T - r/c$. A full discussion of the scattered intensity is given by Kimble and Mandel (1975a). Now the time dependence of the probability $P(\omega, r, T)$ has a double origin. There is a non-stationary emission process and a superimposed T dependence associated with the finite time for detection. For the steady-state probability $P_{ss}(\omega, r, T)$ arising from equation (4.8) in the limit $\hat{t} \rightarrow \infty$ only the second consideration remains. We find

$$P_{ss}(\omega, r, T) = P_{ss}^{\omega_0}(\omega, r, T) + P_{ss}^{\omega_{21}}(\omega, r, T) + P_{ss}^{\omega_{12}}(\omega, r, T) \quad (4.22)$$

with

$$\begin{aligned}P_{ss}^{\omega_0}(\omega, r, T) &= \frac{I_0(r)}{2}\left(\frac{\frac{1}{2}\gamma}{(\frac{1}{2}\gamma)^2 + (\omega - \omega_0)^2}\hat{T} - \frac{(\frac{1}{2}\gamma)^2 - (\omega - \omega_0)^2}{[(\frac{1}{2}\gamma)^2 + (\omega - \omega_0)^2]^2}\right. \\ &\quad \times [1 - e^{-\gamma\hat{T}/2}\cos(\omega - \omega_0)\hat{T}] - \frac{\gamma(\omega - \omega_0)}{[(\frac{1}{2}\gamma)^2 + (\omega - \omega_0)^2]^2} \\ &\quad \left.\times e^{-\gamma\hat{T}/2}\sin(\omega - \omega_0)\hat{T}\right) \quad (4.23)\end{aligned}$$

$$\begin{aligned}P_{ss}^{\omega_{21}}(\omega, r, T) &= \frac{I_0(r)}{4}\left(\frac{\frac{3}{4}\gamma}{(\frac{3}{4}\gamma)^2 + (\omega - \omega_{21})^2}\hat{T} - \frac{(\frac{3}{4}\gamma)^2 - (\omega - \omega_{21})^2}{[(\frac{3}{4}\gamma)^2 + (\omega - \omega_{21})^2]^2}\right. \\ &\quad \times [1 - e^{-3\gamma\hat{T}/4}\cos(\omega - \omega_{21})\hat{T}] - \frac{\frac{3}{4}\gamma(\omega - \omega_{21})}{[(\frac{3}{4}\gamma)^2 - (\omega - \omega_{21})^2]^2} \\ &\quad \left.\times e^{-3\gamma\hat{T}/4}\sin(\omega - \omega_{21})\hat{T}\right) \quad (4.24)\end{aligned}$$

$$\begin{aligned}P_{ss}^{\omega_{12}}(\omega, r, T) &= \frac{I_0(r)}{4}\left(\frac{\frac{3}{4}\gamma}{(\frac{3}{4}\gamma)^2 + (\omega - \omega_{12})^2}\hat{T} - \frac{(\frac{3}{4}\gamma)^2 - (\omega - \omega_{12})^2}{[(\frac{3}{4}\gamma)^2 + (\omega - \omega_{12})^2]^2}\right. \\ &\quad \times [1 - e^{-3\gamma\hat{T}/4}\cos(\omega - \omega_{12})\hat{T}] - \frac{\frac{3}{4}\gamma(\omega - \omega_{12})}{[(\frac{3}{4}\gamma)^2 - (\omega - \omega_{12})^2]^2} \\ &\quad \left.\times e^{-3\gamma\hat{T}/4}\sin(\omega - \omega_{12})\hat{T}\right). \quad (4.25)\end{aligned}$$

These results may be combined with equations (4.20) and (4.21), where we retain the first terms only, to give the spectrum obtained from a saturated atom after a finite time T . Clearly for $T \rightarrow \infty$ we regain equation (4.16). For the complete non-stationary correlation function (4.18) the spectrum is extremely complicated. We find it possible however to make a general prescription in the form

$$I(\omega, r, T) = I(\omega, r, \infty) \frac{I(r, T)}{I(r, \infty)} + I'(\omega, r, T). \quad (4.26)$$

The first term is simply the three-peaked spectrum of the steady state modulated by the scattered intensity. The term $I'(\omega, r, T)$ which through the transient region modifies the shape of these three peaks contains zero integrated intensity. A full expression of $I'(\omega, r, T)$ is given in the appendix.

Of course no restriction on the number of photon emissions has been made to this stage and equation (4.16) agrees with results by Mollow (1969, 1975a, b), Swain (1975) and Smithers and Freedhoff (1975). We will proceed now to show how a one-photon approximation may be introduced into our formalism demonstrating the inadequacy of omitting photon cascades.

5. One-photon approximation

Taking matrix elements diagonal in the field only, the master equation (2.9) reads

$$\frac{d\rho_{n,\eta;n,\xi}}{dt} = \frac{-i}{\hbar} (E_n^\eta - E_n^\xi) \rho_{n,\eta;n,\xi} + (-1)^{\eta+\xi} \frac{\gamma}{4} \sum_{u,v} \rho_{n+1,\mu;n+1,v} - \frac{\gamma}{4} \sum_v (\rho_{n,\eta;n,v} + \rho_{n,v;n,\xi}). \quad (5.1)$$

Let us now take the incident field initially in a Fock state with N photons and the atom excited. All other modes are initially in the vacuum and therefore the possible subsequent states of the coupled field plus atom are $|N-n, +\rangle$, $|N-n+1, -\rangle$, n varying from 0 to N . Correspondingly in the H_S representation we have states $|E_n^2\rangle$ and $|E_n^1\rangle$ for which from equation (5.1) we may write a set of $N+1$ equations. As $\rho_{N+1,\eta;N+1,\xi}$ must remain zero at all times, there are N coupled equations, formally the same as equation (5.1), while for $\rho_{N,\eta;N,\xi}$

$$\frac{d\rho_{N,\eta;N,\xi}}{dt} = \frac{-i}{\hbar} (E_N^\eta - E_N^\xi) \rho_{N,\eta;N,\xi} - \frac{\gamma}{4} \sum_v (\rho_{N,\eta;N,v} + \rho_{N,v;N,\xi}) \quad (5.2)$$

which yields

$$\frac{d}{dt} \begin{pmatrix} \rho_{N,2;N,2} \\ \rho_{N,1;N,1} \\ \rho_{N,2;N,1} \\ \rho_{N,1;N,2} \end{pmatrix} = \begin{pmatrix} -\frac{1}{2}\gamma & 0 & -\frac{1}{4}\gamma & -\frac{1}{4}\gamma \\ 0 & -\frac{1}{2}\gamma & -\frac{1}{4}\gamma & -\frac{1}{4}\gamma \\ -\frac{1}{4}\gamma & -\frac{1}{4}\gamma & -(\frac{1}{2}\gamma + 2iN^{1/2}\kappa) & 0 \\ -\frac{1}{4}\gamma & -\frac{1}{4}\gamma & 0 & -(\frac{1}{2}\gamma - 2iN^{1/2}\kappa) \end{pmatrix} \begin{pmatrix} \rho_{N,2;N,2} \\ \rho_{N,1;N,1} \\ \rho_{N,2;N,1} \\ \rho_{N,1;N,2} \end{pmatrix}. \quad (5.3)$$

Solving this system by the method of §3 we find

$$\rho_{N,2;N,2}(t) = \rho_{N,1;N,1}(t) = -\frac{1}{2} e^{-\gamma t/2} \left[\left(\frac{2N^{1/2}\kappa}{\Omega'} \right)^2 - \left(\frac{\frac{1}{2}\gamma}{\Omega'} \right)^2 \cosh \Omega' t + \frac{\frac{1}{2}\gamma}{\Omega'} \sinh \Omega' t \right] \quad (5.4)$$

$$\rho_{N,2:N,1}(t) = -\frac{1}{2} e^{-\gamma t/2} \left[\frac{2iN^{1/2}\kappa}{\Omega'} \frac{\frac{1}{2}\gamma}{\Omega'} - \left(1 + \frac{2iN^{1/2}\kappa}{\Omega'} \frac{\frac{1}{2}\gamma}{\Omega'} \right) \cosh \Omega' t \right. \\ \left. + \left(\frac{\frac{1}{2}\gamma}{\Omega'} + \frac{2iN^{1/2}\kappa}{\Omega'} \right) \sinh \Omega' t \right] \quad (5.5)$$

$$\rho_{N,1:N,2}(t) = \frac{1}{2} e^{-\gamma t/2} \left[\frac{2iN^{1/2}\kappa}{\Omega'} \frac{\frac{1}{2}\gamma}{\Omega'} + \left(1 - \frac{2iN^{1/2}\kappa}{\Omega'} \frac{\frac{1}{2}\gamma}{\Omega'} \right) \cosh \Omega' t \right. \\ \left. - \left(\frac{\frac{1}{2}\gamma}{\Omega'} - \frac{2iN^{1/2}\kappa}{\Omega'} \right) \sinh \Omega' t \right] \quad (5.6)$$

where

$$\Omega' = [(\frac{1}{2}\gamma)^2 - 4N\kappa^2]^{1/2}. \quad (5.7)$$

We will concern ourselves from here only with the limit of intense illumination

$$4N\kappa^2 \gg (\frac{1}{2}\gamma)^2. \quad (5.8)$$

Here

$$\rho_{N,2:N,2}(t) = \rho_{N,1:N,1}(t) = \frac{1}{2} e^{-\gamma t/2} \\ \rho_{N,2:N,1}(t) = \frac{1}{2} \exp[-(\frac{1}{2}\gamma + 2iN^{1/2}\kappa)t] \\ \rho_{N,1:N,2}(t) = \frac{1}{2} \exp[-(\frac{1}{2}\gamma - 2iN^{1/2}\kappa)t] \quad (5.9)$$

which gives for the probability that the coupled atom plus field remains in its initial state

$$\rho_{N,+;N,+}(t) = e^{-\gamma t/2} \cos^2 N^{1/2}\kappa t. \quad (5.10)$$

This is just the result obtained by Stroud (1971) in a one-photon approximation. This is not however the probability that the first photon emission has not taken place, as stated by Stroud. We must recognize the possibility with an initially excited atom of realizing the state $|N+1, -\rangle$ without emission to the vacuum. We find from equation (5.9)

$$\rho_{N,+;N,+}(t) + \rho_{N+1,-;N+1,-}(t) = e^{-\gamma t/2}. \quad (5.11)$$

The probability for no emission therefore decays exponentially, without modulation. This is missed in Stroud's analysis since his basis does not include the state $|N+1, -\rangle$.

We now calculate the first-order correlation function and spectrum in a one-photon approximation by restricting ourselves to the reduced set of basis states $|E_N^2\rangle$, $|E_N^1\rangle$ and $|E_{N-1}^2\rangle$, $|E_{N-1}^1\rangle$ corresponding to the possible states involved in the scattering of the first photon. The calculation proceeds essentially as before with equation (2.28) replaced by

$$\Sigma_{\eta,\xi}(\tau) = \langle E_N^\eta | \Sigma(\tau) | E_{N-1}^\xi \rangle. \quad (5.12)$$

This obeys an equation formally equivalent to equation (5.2). After a transformation $\Sigma_{\eta\xi}(\tau) = \hat{\Sigma}_{\eta\xi}(\tau) e^{-i\omega_0\tau}$ the matrix equations of these matrix elements reduce to a form equivalent to equations (5.3).

Now in equation (5.9) there is no steady state as the matrix elements decay eventually to those of lower photon number. In the knowledge that a steady-state spectrum does arise however, in the one-photon approximation we see it maintained by a series of independent emissions from the atomic excited state, each followed by

pumping back to the excited state by the incident field. The spectrum is then built up from a large number of identical contributions arising in separate transitions. In solving the equations for $\Sigma_{je}(\tau)$ we then take for the initial conditions required by equations (2.23) and (5.11) those defined by equation (5.9) for $\hat{t} = 0$. We find from equations (2.14) and (2.26)

$$G_{ss}^{(1)}(\tau) = \frac{1}{2} I_0(r) (\exp[-(\frac{1}{2}\gamma + i\omega_0)\tau] + \frac{1}{2} \exp\{-[\frac{1}{2}\gamma + i(\omega_0 + 2N^{1/2}\kappa)\tau]\} \\ + \frac{1}{2} \exp\{-[\frac{1}{2}\gamma + i(\omega_0 - 2N^{1/2}\kappa)\tau]\}). \quad (5.13)$$

Substituting into equation (4.14) gives

$$I(\omega, r, \infty) = \frac{I_0(r)}{2\pi} \left(\frac{\frac{1}{2}\gamma}{(\frac{1}{2}\gamma)^2 + (\omega - \omega_0)^2} + \frac{1}{2} \frac{\frac{1}{2}\gamma}{(\frac{1}{2}\gamma)^2 + [\omega - (\omega_0 + 2N^{1/2}\kappa)]^2} \right. \\ \left. + \frac{1}{2} \frac{\frac{1}{2}\gamma}{(\frac{1}{2}\gamma)^2 + [\omega - (\omega_0 - 2N^{1/2}\kappa)]^2} \right). \quad (5.14)$$

This spectrum is the same as that obtained by Stroud (1971) with the absence of his linewidth narrowing (for the origin of this feature see Mollow 1975). In comparison with equation (4.16) (neglecting the coherent scattering term) we see then the inadequacy of the one-photon approximation in predicting linewidths and peak heights. In equation (4.16) sidebands are broadened from the central peak by a factor of 3:2 and peak heights are in the ratio of 3:1. The one-photon approximation omits the sideband broadening and gives the ratio of peak heights as 2:1 (Carmichael and Walls 1975a). With the scattered intensity given by $G_{ss}^{(1)}(0)$ we might note also the reduction of intensity by a factor of $\frac{1}{2}$ in equation (4.16) as compared to equation (5.15). This arises in the depletion of the initial scattered intensity to half its value at saturation. From equation (4.8) we have

$$G^{(1)}(r, t; r, t) = \frac{1}{2} I_0(r) (1 + e^{-3\gamma\hat{t}/4} \cos 2\hbar^{1/2}\kappa\hat{t}). \quad (5.15)$$

We note in conclusion that while we have restricted ourselves here for the most part to the limit (equation (5.8)), for arbitrary intensities we will obtain the general features of the previous section with splitting of the spectrum at a threshold. Clearly from equation (5.7) however, the one-photon approximation sets this threshold at

$$\frac{1}{2}\gamma = 2N^{1/2}\kappa \quad (5.16)$$

indicating twice the intensity required by equation (4.12).

6. The second-order correlation function

We turn in this final section to the second-order correlation function and intensity correlations. Calculation of this function is readily available in our formalism. With equations (2.17) and (2.27) we require only the solution to equations (2.31). This follows again from the scheme outlined for atomic dynamics, and defining

$$\tilde{\Pi}(\tau) = \frac{1}{2} \begin{pmatrix} \Pi_{22} + \Pi_{11} \\ \Pi_{22} - \Pi_{11} \\ \Pi_{21} + \Pi_{12} \\ \Pi_{21} - \Pi_{12} \end{pmatrix} \quad (6.1)$$

we may write

$$\tilde{\Pi}(\tau) = \mathbf{S} \exp(\Lambda\tau) \mathbf{S}^{-1} \tilde{\Pi}(0) \quad (6.2)$$

where $\mathbf{S}$ and Λ are as defined in equations (3.3) and (3.4). From equation (2.24) we find

$$\tilde{\Pi}(0) = \frac{1}{2} \begin{pmatrix} \rho_{+,+}(\hat{t}) \\ 0 \\ -\rho_{+,+}(\hat{t}) \\ 0 \end{pmatrix}. \quad (6.3)$$

We will concern ourselves only with the steady state and therefore from equations (3.6) and (3.10) the initial condition is specified by

$$\rho_{+,+}^{ss} = \frac{4\bar{n}\kappa^2}{\gamma^2 + 8\bar{n}\kappa^2}. \quad (6.4)$$

Substitution in equation (6.2) yields

$$G_{ss}^{(2)}(\tau) = G_{ss}^{(1)}(0)^2 \left[1 - e^{-3\gamma\tau/4} \left(\cosh \Omega\tau + \frac{3\gamma}{\Omega} \sinh \Omega\tau \right) \right] \quad (6.5)$$

where $G_{ss}^{(1)}(0)$ is simply the steady-state intensity

$$G_{ss}^{(1)}(0) = I_0(r) \frac{4\bar{n}\kappa^2}{\gamma^2 + 8\bar{n}\kappa^2}. \quad (6.6)$$

Now the familiar demonstration of second-order correlation effects is, of course, the photon-bunching phenomenon in the Hanbury Brown and Twiss experiment (Hanbury Brown and Twiss 1956). In contrast to this there are also fields for which photons tend to be separated, on the average producing second-order correlations which fall below $G_{ss}^{(1)}(0)^2$ as τ approaches zero. Such an effect has been termed photon antibunching and arises for example in parametric subharmonic generation (Stoler 1974). Turning then to equation (6.5) we find just this behaviour where for $\tau = 0$, the second-order correlation function vanishes. The interpretation of this is simple. It of course depends solely on the quantum nature of the scattering. Consider a photon detected at a position r and time $\hat{t}$. This then serves to identify the atom in its lower state at time t so that we may view this emission as preparing the atom in its ground state. Knowing that any subsequent emission must begin with an excited atom, a delay corresponding to the time taken to regain this excited condition is naturally expected. We ask therefore—what is the probability for finding an initially unexcited atom in its upper state? The answer is given by equation (3.6) and is just the expression (6.5) for $G_{ss}^{(2)}(\tau)$. It has been pointed out by Cohen-Tannoudji (1976) who independently suggested the use of photon-correlation techniques in resonance fluorescence experiments that this provides an example of the principle of reduction of the wavepacket in quantum mechanics.

In the weak field limit, equation (6.5) takes the form displayed in figure 1:

$$G_{ss}^{(2)}(\tau) = G_{ss}^{(1)}(0)^2 (1 - e^{-\gamma\tau/2})^2. \quad (6.7)$$

Here this antibunching phenomenon is particularly significant since it provides a purely QED prediction in a region which is otherwise adequately described in a semiclassical treatment. Measurement of $G_{ss}^{(2)}(\tau)$ therefore presents the possibilities for a further test of

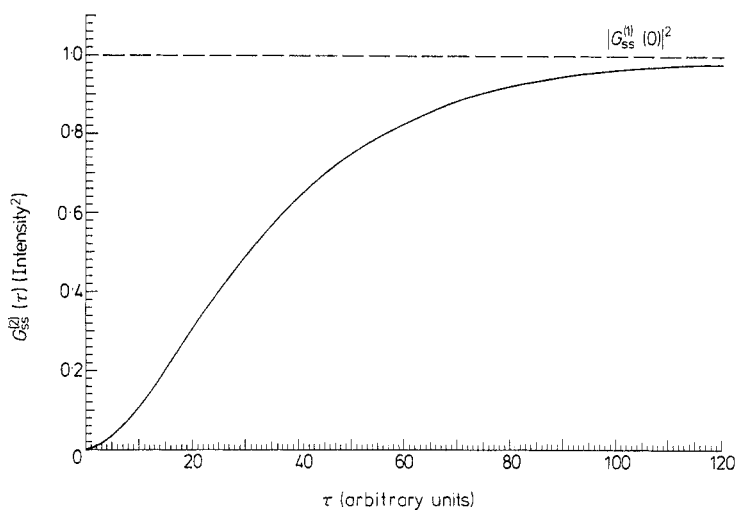


Figure 1. Second-order correlation function for light scattered by a single atom, $2\bar{n}^{1/2}\kappa \ll \frac{1}{4}\gamma$.

QED. For strong illumination the correlation function is presented in figure 2 and corresponds to the form:

$$G_{ss}^{(2)}(\tau) = G_{ss}^{(1)}(0)^2 (1 - e^{-3\gamma\tau/4} \cos 2\bar{n}^{1/2}\kappa\tau). \quad (6.8)$$

Here we have both photon bunching and antibunching displayed in the one situation. This of course corresponds to the oscillation of the probability for excitation through the transient regime between values above and below that attained at saturation.

Let us now consider $G_{ss}^{(2)}(\tau)$ from another perspective: that which sees in it a possible source for the experimental measurement of spectral parameters. We are concerned then particularly with the form (6.8) which is associated with the three-peaked spectrum given in equation (4.16). We see directly that contained in this function is information on both the peak widths and the splitting frequency. With the measurement of

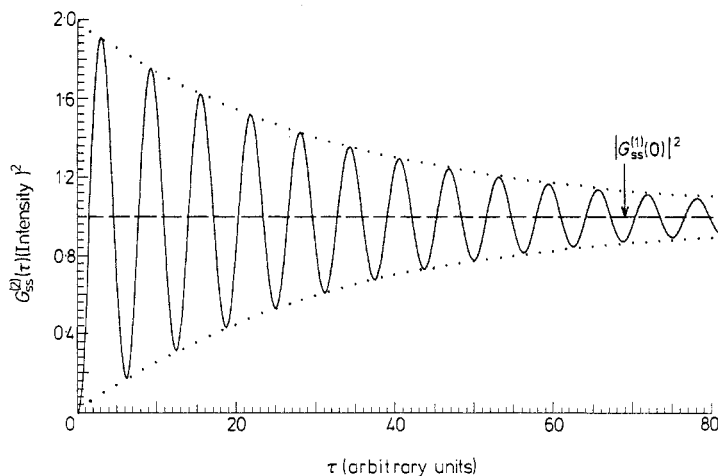


Figure 2. Second-order correlation function for light scattered by a single atom, $2\bar{n}^{1/2}\kappa \gg \frac{1}{4}\gamma$.

$G_{ss}^{(2)}(\tau)$ we can then extract spectral detail which has to this stage been unavailable (Carmichael and Walls 1975b, 1976).

Now equation (6.5) applies to the scattering from a single atom repeatedly absorbing and re-emitting photons from the incident field. There is clearly then going to be a major problem obtaining sufficient scattered intensity in any attempt to measure this correlation function. In view of this fact we might usefully consider the simultaneous illumination of many atoms. This corresponds to the experiments of Schuda *et al* (1974), Walther (1975), Wu *et al* (1975) and Hartig *et al* (1976), where an atomic beam is arranged to cross a laser field so that many atoms experience irradiation at one time. The scattered field $E_s(r, t)$ becomes the sum of fields $E_{s_k}(r, t)$ arising from individual atoms

$$E_s(r, t) = \sum_k E_{s_k}(r, t). \quad (6.9)$$

Now these scattering centres enter the laser field at random times and may be taken to act independently. This means that the component of $G_{ss}^{(2)}(\tau)$ corresponding to the second-order correlations for individual atoms is swamped in the limit of many atoms by that corresponding to the product of first-order correlations. This reflects the introduction of Gaussian statistics which follow from the central limit theorem. We find a result which holds generally for Gaussian signals (Glauber 1963):

$$G^{(2)}(r, t; r, t + \tau) = G^{(1)}(r, t; r, t)^2 + |G^{(1)}(r, t; r, t + \tau)|^2. \quad (6.10)$$

We may take the illuminated atoms in their saturated state and then the summed field will be stationary and the first-order correlation function simply proportional to that for a single atom. For weak and strong illumination respectively we may therefore write from equations (4.13) and (4.14)

$$|G_{ss}^{(1)}(\tau)| \propto I_0(r) \frac{4\bar{n}\kappa^2}{\gamma^2 + 8\bar{n}\kappa^2} \quad (6.11)$$

and

$$|G_{ss}^{(1)}(\tau)| \propto \frac{I_0(r)}{2} \frac{1}{2} (e^{-\gamma\tau/2} + e^{-3\gamma\tau/4} \cos 2\bar{n}^{1/2}\kappa\tau). \quad (6.12)$$

The property of photon antibunching is lost here as we expect, nonetheless equation (6.12) still contains the spectral information hoped for. Indeed it bears possibilities

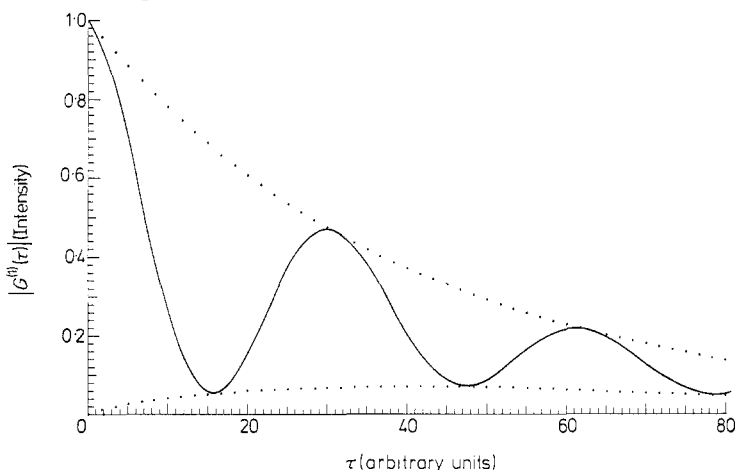


Figure 3. Second-order correlation function for light scattered by many atoms, $2\bar{n}^{1/2}\kappa : \frac{1}{4}\gamma = 20:1$. Sideband displacement 10 times natural linewidth.

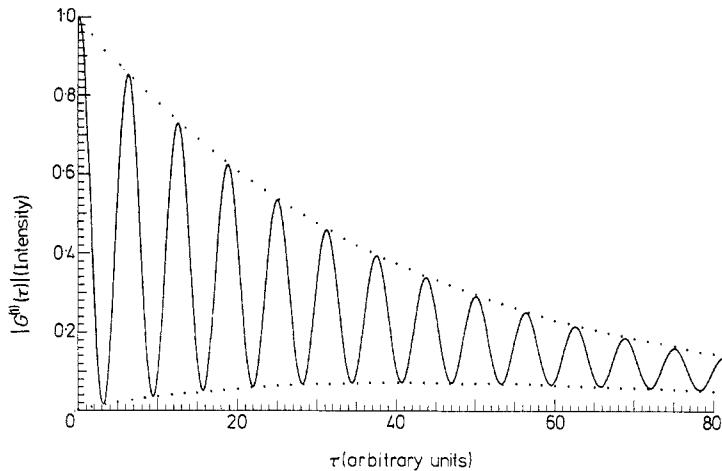


Figure 4. Second-order correlation function for light scattered by many atoms, $2\bar{n}^{1/2}\kappa:\frac{1}{4}\gamma = 100:1$. Sideband displacement 50 times natural linewidth.

over and above those found in equation (6.8). In figures 3 and 4 clearly the addition and subtraction of the upper and lower envelopes makes available both the curves $e^{-\gamma\tau/2}$ and $e^{-3\gamma\tau/4}$. Here then is the potential for a direct test of the predicted ratio, 3:2, for sideband broadening.

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Appendix

We present here the full form for the term $I'(\omega, r, T)$ in equation (4.26). From equation (4.2) we write

$$I'(\omega, r, T) = \frac{1}{2\pi} \frac{I(r, T)}{\int_{r/c}^T dt I(r, t)} P'(\omega, r, T) \quad (\text{A.1})$$

with the intensity and integrated intensity given by equations (4.20) and (4.21). $P'(\omega, r, T)$ is conveniently written as the sum of four terms

$$P'(\omega, r, T) = -\frac{1}{2} I_0(r) \sum_{k=1}^4 \mathcal{P}_k(\omega, T). \quad (\text{A.2})$$

We have

$$\begin{aligned} \mathcal{P}_1(\omega, T) = & \frac{1}{2} \left(\frac{\omega - \omega_{21}}{(\frac{3}{4}\gamma)^2 + (\omega - \omega_{21})^2} - \frac{\omega - \omega_{12}}{(\frac{3}{4}\gamma)^2 + (\omega - \omega_{12})^2} \right) \frac{2\bar{n}^{1/2}\kappa}{(\frac{3}{4}\gamma)^2 + 4\bar{n}\kappa^2} \\ & \times (1 - e^{-3\gamma\hat{T}/4} \cos 2\bar{n}^{1/2}\kappa\hat{T}) - \frac{1}{2} \left(\frac{\omega - \omega_{21}}{(\frac{3}{4}\gamma)^2 + (\omega - \omega_{21})^2} \right. \\ & \left. - \frac{\omega - \omega_{12}}{(\frac{3}{4}\gamma)^2 + (\omega - \omega_{12})^2} \right) \frac{\frac{3}{4}\gamma}{(\frac{3}{4}\gamma)^2 + 4\bar{n}\kappa^2} e^{-3\gamma\hat{T}/4} \sin 2\bar{n}^{1/2}\kappa\hat{T} \end{aligned} \quad (\text{A.3})$$

$$\begin{aligned}
\mathcal{P}_2(\omega, T) = & \left(\frac{\frac{1}{2}\gamma}{(\frac{1}{2}\gamma)^2 + (\omega - \omega_0)^2} \frac{\frac{1}{2}\gamma}{(\frac{1}{2}\gamma)^2 + (\omega - \omega_0)^2} - \frac{\omega - \omega_0}{(\frac{1}{2}\gamma)^2 + (\omega - \omega_0)^2} \right. \\
& \times \left. \frac{\omega - \omega_0}{(\frac{1}{2}\gamma)^2 - (\omega - \omega_0)^2} \right) [1 - e^{-\gamma\hat{T}/2} \cos(\omega - \omega_0)\hat{T}] \\
& + \left(\frac{\frac{1}{2}\gamma}{(\frac{1}{2}\gamma)^2 + (\omega - \omega_0)^2} \frac{\omega - \omega_0}{(\frac{1}{2}\gamma)^2 + (\omega - \omega_0)^2} + \frac{\frac{1}{2}\gamma}{(\frac{1}{2}\gamma)^2 + (\omega - \omega_0)^2} \right. \\
& \times \left. \frac{\omega - \omega_0}{(\frac{1}{2}\gamma)^2 + (\omega - \omega_0)^2} \right) e^{-\gamma\hat{T}/2} \sin(\omega - \omega_0)\hat{T} + \frac{1}{2} \left(\frac{\frac{3}{2}\gamma}{(\frac{3}{2}\gamma)^2 + (\omega - \omega_0)^2} \right. \\
& \times \left. \frac{\frac{3}{4}\gamma}{(\frac{3}{4}\gamma)^2 + (\omega - \omega_{21})^2} - \frac{\omega - \omega_{21}}{(\frac{3}{4}\gamma)^2 + (\omega - \omega_{21})^2} \frac{\omega - \omega_0}{(\frac{3}{2}\gamma)^2 + (\omega - \omega_0)^2} \right) \\
& \times [1 - e^{-3\gamma\hat{T}/2} \cos(\omega - \omega_0)\hat{T}] + \frac{1}{2} \left(\frac{\frac{3}{2}\gamma}{(\frac{3}{2}\gamma)^2 + (\omega - \omega_0)^2} \right. \\
& \times \left. \frac{\omega - \omega_{21}}{(\frac{3}{4}\gamma)^2 + (\omega - \omega_{21})^2} + \frac{\frac{3}{4}\gamma}{(\frac{3}{4}\gamma)^2 + (\omega - \omega_{21})^2} \frac{\omega - \omega_0}{(\frac{3}{2}\gamma)^2 + (\omega - \omega_0)^2} \right) \\
& \times e^{-3\gamma\hat{T}/2} \sin(\omega - \omega_0)\hat{T} + \frac{1}{2} \left(\frac{\frac{3}{2}\gamma}{(\frac{3}{2}\gamma)^2 + (\omega - \omega_0)^2} \frac{\frac{3}{4}\gamma}{(\frac{3}{4}\gamma)^2 + (\omega - \omega_{12})^2} \right. \\
& \times \left. \frac{\omega - \omega_{12}}{(\frac{3}{4}\gamma)^2 + (\omega - \omega_{12})^2} \frac{\omega - \omega_0}{(\frac{3}{2}\gamma)^2 + (\omega - \omega_0)^2} \right) [1 - e^{-3\gamma\hat{T}/2} \cos(\omega - \omega_0)\hat{T}] \\
& + \frac{1}{2} \left(\frac{\frac{3}{2}\gamma}{(\frac{3}{2}\gamma)^2 + (\omega - \omega_0)^2} \frac{\omega - \omega_{12}}{(\frac{3}{4}\gamma)^2 + (\omega - \omega_{12})^2} + \frac{\frac{3}{4}\gamma}{(\frac{3}{4}\gamma)^2 + (\omega - \omega_{12})^2} \right. \\
& \times \left. \frac{\omega - \omega_0}{(\frac{3}{2}\gamma)^2 + (\omega - \omega_0)^2} \right) e^{-3\gamma\hat{T}/2} \sin(\omega - \omega_0)\hat{T}. \tag{A.4}
\end{aligned}$$

$$\begin{aligned}
\mathcal{P}_3(\omega, T) = & \frac{1}{2} \left(\frac{\frac{3}{4}\gamma}{(\frac{3}{4}\gamma)^2 + (\omega - \omega_{21})^2} \frac{\frac{3}{4}\gamma}{(\frac{3}{4}\gamma)^2 + (\omega - \omega_{21})^2} - \frac{\omega - \omega_{21}}{(\frac{3}{4}\gamma)^2 + (\omega - \omega_{21})^2} \right. \\
& \times \left. \frac{\omega - \omega_{21}}{(\frac{3}{4}\gamma)^2 + (\omega - \omega_{21})^2} \right) [1 - e^{-3\gamma\hat{T}/4} \cos(\omega - \omega_{21})\hat{T}] \\
& + \frac{1}{2} \left(\frac{\frac{3}{4}\gamma}{(\frac{3}{4}\gamma)^2 + (\omega - \omega_{21})^2} \frac{\omega - \omega_{21}}{(\frac{3}{4}\gamma)^2 + (\omega - \omega_{21})^2} + \frac{\frac{3}{4}\gamma}{(\frac{3}{4}\gamma)^2 + (\omega - \omega_{21})^2} \right. \\
& \times \left. \frac{\omega - \omega_{21}}{(\frac{3}{4}\gamma)^2 + (\omega - \omega_{21})^2} \right) e^{-3\gamma\hat{T}/4} \sin(\omega - \omega_{21})\hat{T} \\
& + \frac{1}{2} \left(\frac{\frac{1}{2}\gamma}{(\frac{1}{2}\gamma)^2 + (\omega - \omega_0)^2} \frac{\frac{5}{4}\gamma}{(\frac{5}{4}\gamma)^2 + (\omega - \omega_{21})^2} - \frac{\omega - \omega_{21}}{(\frac{5}{4}\gamma)^2 + (\omega - \omega_{21})^2} \right. \\
& \times \left. \frac{\omega - \omega_0}{(\frac{1}{2}\gamma)^2 + (\omega - \omega_0)^2} \right) [1 - e^{-5\gamma\hat{T}/4} \cos(\omega - \omega_{21})\hat{T}] \\
& + \frac{1}{2} \left(\frac{\frac{1}{2}\gamma}{(\frac{1}{2}\gamma)^2 + (\omega - \omega_0)^2} \frac{\omega - \omega_{21}}{(\frac{5}{4}\gamma)^2 + (\omega - \omega_{21})^2} + \frac{\frac{5}{4}\gamma}{(\frac{5}{4}\gamma)^2 + (\omega - \omega_{21})^2} \right. \\
& \times \left. \frac{\omega - \omega_0}{(\frac{1}{2}\gamma)^2 + (\omega - \omega_0)^2} \right) e^{-5\gamma\hat{T}/4} \sin(\omega - \omega_{21})\hat{T}. \tag{A.5}
\end{aligned}$$

$$\begin{aligned}
\mathcal{P}_4(\omega, T) = & \frac{1}{2} \left(\frac{\frac{3}{4}\gamma}{(\frac{3}{4}\gamma)^2 + (\omega - \omega_{12})^2} \frac{\frac{3}{4}\gamma}{(\frac{3}{4}\gamma)^2 + (\omega - \omega_{12})^2} - \frac{\omega - \omega_{12}}{(\frac{3}{4}\gamma)^2 + (\omega - \omega_{12})^2} \right. \\
& \times \left. \frac{\omega - \omega_{12}}{(\frac{3}{4}\gamma)^2 + (\omega - \omega_{12})^2} \right) [1 - e^{-3\gamma\hat{T}/4} \cos(\omega - \omega_{12})\hat{T}] \\
& + \frac{1}{2} \left(\frac{\frac{3}{4}\gamma}{(\frac{3}{4}\gamma)^2 + (\omega - \omega_{12})^2} \frac{\omega - \omega_{12}}{(\frac{3}{4}\gamma)^2 + (\omega - \omega_{12})^2} + \frac{\frac{3}{4}\gamma}{(\frac{3}{4}\gamma)^2 + (\omega - \omega_{12})^2} \right. \\
& \times \left. \frac{\omega - \omega_{12}}{(\frac{3}{4}\gamma)^2 + (\omega - \omega_{12})^2} \right) e^{-3\gamma\hat{T}/4} \sin(\omega - \omega_{12})\hat{T} \\
& + \frac{1}{2} \left(\frac{\frac{1}{2}\gamma}{(\frac{1}{2}\gamma)^2 + (\omega - \omega_0)^2} \frac{\frac{5}{4}\gamma}{(\frac{5}{4}\gamma)^2 + (\omega - \omega_{12})^2} - \frac{\omega - \omega_{12}}{(\frac{5}{4}\gamma)^2 + (\omega - \omega_{12})^2} \right. \\
& \times \left. \frac{\omega - \omega_0}{(\frac{1}{2}\gamma)^2 + (\omega - \omega_0)^2} \right) [1 - e^{-5\gamma\hat{T}/4} \cos(\omega - \omega_{12})\hat{T}] \\
& + \frac{1}{2} \left(\frac{\frac{1}{2}\gamma}{(\frac{1}{2}\gamma)^2 + (\omega - \omega_0)^2} \frac{\omega - \omega_{12}}{(\frac{5}{4}\gamma)^2 + (\omega - \omega_{12})^2} + \frac{\frac{5}{4}\gamma}{(\frac{5}{4}\gamma)^2 + (\omega - \omega_{12})^2} \right. \\
& \times \left. \frac{\omega - \omega_0}{(\frac{1}{2}\gamma)^2 + (\omega - \omega_0)^2} \right) e^{-5\gamma\hat{T}/4} \sin(\omega - \omega_{12})\hat{T}. \tag{A.6}
\end{aligned}$$

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